

CLAIMS

- 1        1. (Currently Amended) A process of preparing ortho substituted phenylamines comprising  
2        contacting phenylhydroxylamine, optionally substituted with at least one inert substituent,  
3        with a nucleophilic reagent in the presence of a manganese oxide at a temperature between  
4        about 10° C and about 170° C and a pressure from subatmospheric to superatmospheric such  
5        that an ortho substituted phenylamine, optionally correspondingly substituted with at least  
6        one inert substituent, is predominantly formed.
  
- 1        2. (Original) The process of claim 1 wherein the phenylhydroxylamine is unsubstituted  
2        phenylhydroxylamine.
  
- 1        3. (Original) The process of claim 1 wherein the phenylhydroxylamine is substituted with at  
2        least one member selected from the group consisting of C<sub>1</sub>-C<sub>10</sub> alkyl, C<sub>6</sub>-C<sub>10</sub> aryl, and C<sub>6</sub>-C<sub>10</sub>  
3        alkaryl moieties.
  
- 1        4. (Original) The process of claim 1 wherein the nucleophilic reagent is selected from the  
2        group consisting of ammonia, water, C<sub>1</sub>-C<sub>20</sub> aliphatic alcohols, phenols, halides, and amines  
3        having the formula R'<sub>2</sub>NH wherein each R' may independently be a hydrogen, C<sub>1</sub>-C<sub>20</sub>  
4        aliphatic, C<sub>4</sub>-C<sub>8</sub> alicyclic, or C<sub>6</sub>-C<sub>15</sub> aryl or alkaryl moiety.
  
- 1        5. (Original) The process of claim 1 wherein the nucleophilic reagent is an amine represented  
2        by the formula R'<sub>2</sub>NH wherein each R' is independently a hydrogen, C<sub>1</sub>-C<sub>5</sub> alkyl, or C<sub>6</sub>-C<sub>10</sub>  
3        phenyl or alkyl-substituted phenyl moiety.
  
- 1        6. (Original) The process of claim 5 wherein the nucleophilic reagent is aniline.
  
- 1        7. (Original) The process of claim 1 wherein the molar ratio of nucleophilic reagent to  
2        phenylhydroxylamine ranges from about 2 to about 100.
  
- 1        8. (Currently Amended) A process for preparing ortho substituted phenylamines comprising

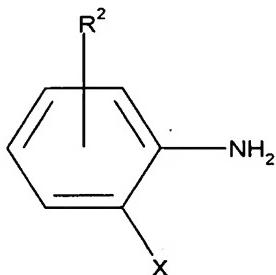
2 contracting phenylhydroxylamine, optionally substituted with at least one inert substituent,  
3 with a nucleophilic reagent, the molar ratio of nucleophilic reagent to phenylhydroxylamine  
4 ranging from about 2 to about 100, the contacting of the phenylhydroxylamine and  
5 nucleophilic reagent being conducted in the absence of oxygen and in the presence of a  
6 catalyst that is a cryptomelane-type manganese oxide Octahedral Molecular Sieve, with a  
7 composition of  $KMn_8O_{16} \cdot nH_2O$  ( $n = 0.5-10$ ) in which said molecular sieve comprises  $MnO_6$   
8 octahedral structural units that are edge and corner shared to form a  $4.6 \times 4.6$  tunnels as a  
9 result of  $2 \times 2$  arrangement of octahedra, in which the potassium ions are present in the  
10 tunnels with a small amount of water and said potassium ions are ion-exchanged by  $H^+$  ions  
11 using nitric acid to obtain the acidic form of said sieve at temperatures ranging from about  
12  $70^\circ C$  to about  $120^\circ C$ , whereby an optionally-substituted ortho substituted phenylamine is  
13 formed in amounts equal to or greater than any concurrently formed para isomer.

1 9. (Original) The process of claim 8 wherein the phenylhydroxylamine is unsubstituted  
2 phenylhydroxylamine.

1 10. (Original) The process of claim 8 wherein the nucleophilic reagent is selected from the  
2 group consisting of ammonia, water,  $C_1-C_{20}$  aliphatic alcohols, phenols, halides, and amines  
3 having the formula  $R'_2NH$  wherein each  $R'$  may independently be a hydrogen,  $C_1-C_{20}$   
4 aliphatic,  $C_4-C_8$  alicyclic, or  $C_6-C_{15}$  aryl or alkaryl moiety.

1 11. (Original) The process of claim 8 wherein the nucleophilic reagent is aniline.

1 12. (Original) The process of claim 8 wherein the ortho substituted phenylamine is  
2 represented by the formula:



9 wherein R<sup>2</sup> is hydrogen or at least one C<sub>1</sub>-C<sub>10</sub> alkyl moiety, and X is selected from hydroxy,  
10 halo, C<sub>1</sub>-C<sub>20</sub> alkoxy, phenoxy, and amino of the formula -NR'<sub>2</sub> wherein each R' is  
11 independently a C<sub>1</sub>-C<sub>20</sub> aliphatic, C<sub>4</sub>-C<sub>8</sub> alicyclic, or C<sub>6</sub>-C<sub>15</sub> aryl or alkaryl moiety.

1 13. (Original) The process of claim 12 wherein X is amino and the ortho substituted  
2 phenylamine is a o-phenylenediamine.

1 14. (Currently Amended) The process of claim 13 wherein ~~the ortho the ortho~~ substituted  
2 phenylamine is o-aminodiphenylamine represented by the formula:

